

# Direct Length Measurement Using X-radiography for the Determination of Acoustic Velocities at High Pressure and Temperature

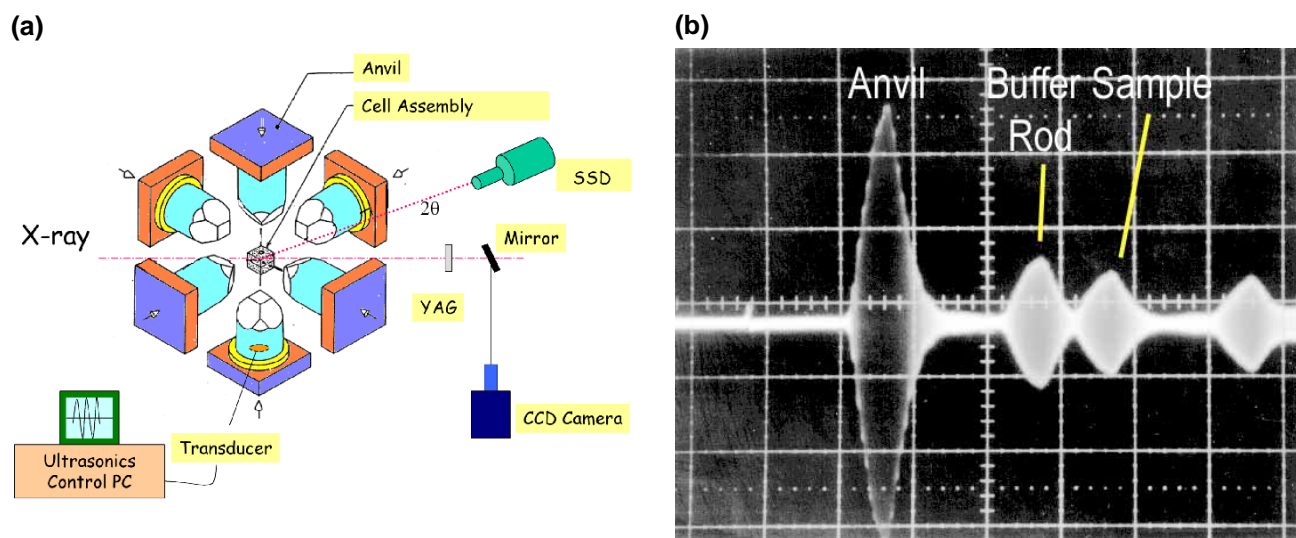
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Physical properties of Earth materials at high pressure and high temperature are of great interest to Earth scientists. These data can be compared with those derived from seismic studies to place constraints on the composition, structure and dynamics of the Earth's interior. Acoustic velocity measurement is an important tool for the determination of elastic properties of solid, liquid and other materials. By measuring the time delay of radio-frequency pulses traveling through a sample and the length of the sample, elastic wave velocities of the sample can be determined precisely. These measurements can be easily accomplished at ambient conditions while the sample lengths can be measured using high precision micrometers. For the study of elastic wave velocities at high pressure and high temperature, however, the sample is commonly enclosed in a high-pressure vessel and direct measurement of sample length becomes difficult. An alternative method is to calculate sample length using previously reported compressibility and thermal expansion data. The error in this approach mainly comes from the reliability of the compressibility/thermal expansion data and the variation of the sample environment (e.g., stress) among different techniques. Recently, a combined ultrasonics

and X-ray diffraction technique has enabled simultaneous measurements of X-ray diffraction and travel time at high pressure and high temperature (e.g., Liebermann and Li, 1998). Using this technique, the sample length at high pressure and temperature is calculated using the unit cell volume derived from X-ray diffraction ( $l/l_0 = (V/V_0)^{1/3}$ ). This method provides precise sample length information only if the sample undergoes elastic deformation during the entire course of the experiments. Large uncertainty can be introduced when plastic deformation occurs during the experiment. Therefore, a direct measurement of sample length at high pressures and high temperatures is very important for obtaining precise velocity measurements.

We have successfully adapted a radiography technique with our simultaneous ultrasonics and X-ray diffraction set-up at the superwiggler beamline X17B1 at the NSLS. This enables us to collect three types of data - ultrasonic travel time, X-ray diffraction, and X-ray imaging of sample length simultaneously in a single high pressure experiment. The ultrasonic interferometry method in this set-up has been described in previous studies (Li et al., 1996, Liebermann and Li, 1998). Briefly, a 250-ton DIA (MA-6) type, cubic anvil high pres-

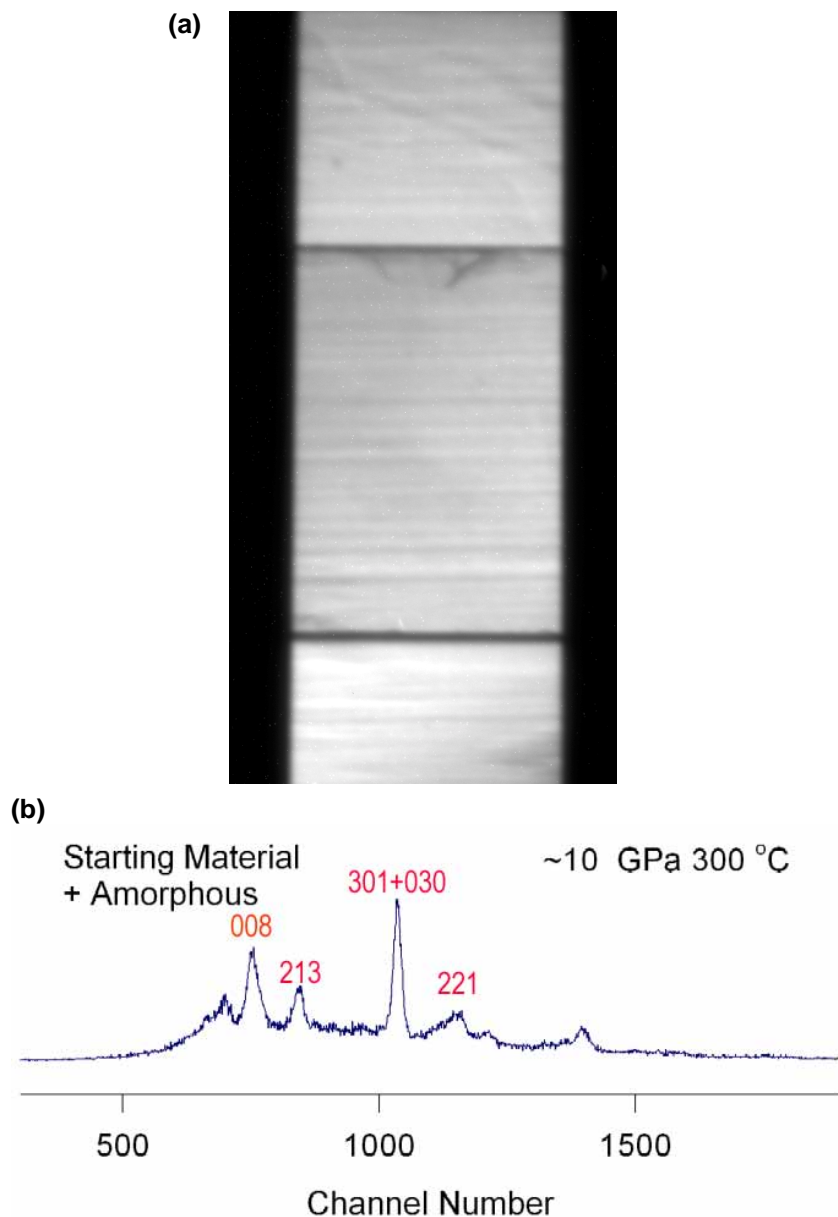


**Figure 1.** (a) Schematic diagram for simultaneous ultrasonics, X-ray diffraction and X-ray imaging studies using a DIA-type high pressure apparatus. (b) Typical acoustic signals showing the reflections from the anvil, buffer rod and sample.

sure apparatus consisting of 4 anvils moving horizontally and two anvils vertically, is used for high pressure generation, (Top and Bottom, see Fig. 1a). On the back of the bottom anvil, a piezoelectric transducer is mounted to generate and receive acoustic signals. The acoustic waves pass through the anvil and propagate into a hybrid buffer rod/sample configuration enclosed in the pressure-transmitting medium. The received acoustic signals are a series of reflections from the interfaces along the wave propagation paths, including the anvil/buffer rod, buffer rod/sample, and sample/post-

sample backing material interfaces. Figure 1b is a typical pattern showing, from left to right, that the first three echoes are from anvil, buffer rod, and sample, respectively. Travel times for P and S waves are measured simultaneously with a dual mode piezoelectric transducer (resonance frequency of P wave at 50 MHz, S waves 30 MHz) using an ultrasonic phase comparison technique (e.g., Niesler and Jackson, 1989). The typical precision for the measured travel time in this configuration is better than 0.5%. Details of the ultrasonic measurements technique can be found in Niesler and Jackson (1989).

The X-ray imaging system consists of a YAG crystal, a mirror, and a CCD camera (Fig. 1a). The YAG crystal is placed in the direct beam path behind the high pressure apparatus. After taking X-ray diffraction data, the front slits are removed to maximize X-ray beam size through the high pressure cell assembly. The fluorescence from the excitation of the X-ray beam passing through the cell assembly and hitting the YAG crystal, is redirected by a mirror and captured by a CCD camera. These images show contrasting brightness from region to region caused by the difference in their X-ray absorption of the materials in the assembly. In this study, gold foil (0.002 mm thick) with a high X-ray absorption coefficient was placed inside the high pressure cell to mark the sample boundaries. These are seen as dark horizontal bands in Figures 2a and 3a. When the sample itself has high X-ray absorption coefficients, no markers are needed. At high pressures, the closure of the first stage tungsten carbide anvils blocks part of the X-ray beam though the cell, only the center portion of the cell assembly is recorded. Calibration is done by comparing the last sample image taken at zero pressure with precise measurement of the recovered sample. All the images collected at sequential P and T conditions are processed to get the intensity values integrated over the sample region along the sample direction. Cross-correlation of the intensities of the sample region between two P-T conditions yields the relative length change from one condition to another. This method provides the power of resolving relative length change of  $10^{-4}$



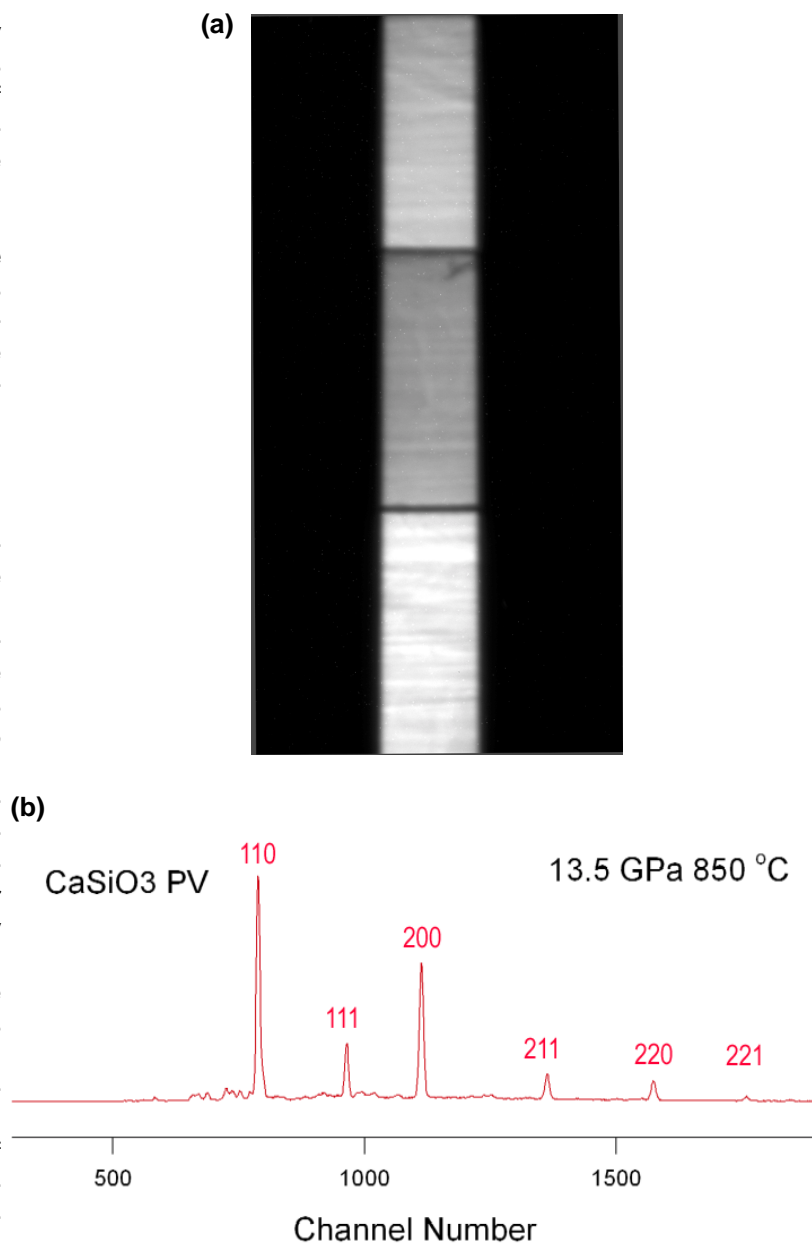
**Figure 2.** Image (2a) and X-ray diffraction pattern (2b) for  $\text{CaSiO}_3$  at 9.6 GPa and room temperature, before the phase transformation,. The two dark lines in this image indicate the position of gold foils placed on both ends of the sample. Triclinic starting material hkl's are shown in red.

(typically less than .25  $\mu\text{m}$  for a 1.0 mm long sample).

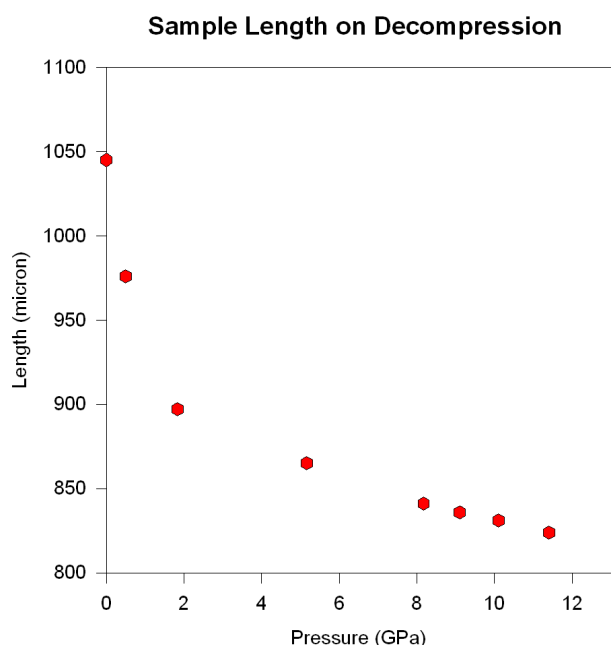
We have utilized this method to study the elastic properties of calcium silicate perovskite, a cubic structured phase of  $\text{CaSiO}_3$  existing in the earth's mantle from the mid-transition zone to the bottom of the lower mantle. Knowing the elastic properties of this phase at mantle conditions is very important in understanding its contribution to the 520 km seismic discontinuity as well as its visibility in the velocity structure of the lower mantle (Karki and Crain, 1998). Since this high pressure phase is not quenchable, i.e., cannot be recovered at ambient conditions, measurements have to be conducted in-situ. Using the method described here, we have successfully synthesized the cubic structured phases and measured elastic wave velocities of P and S waves to 13.5 GPa. In this experiment, a semi-sintered low pressure phase of  $\text{CaSiO}_3$  (Triclinic,  $\rho=2.97 \text{ g/cm}^3$ ) with initial length of 1.850 mm was used as starting material. After reaching the designated pressure, the sample was heated to 850  $^\circ\text{C}$  in 30 minutes. The initial formation and growth of the perovskite phase along heating is monitored by real-time X-ray diffraction. An image of the sample at 9.6 GPa and room temperature is shown in Figure 2a, with the corresponding X-ray diffraction pattern shown in Figure 2b. The equivalent images at 13.5 GPa and 850  $^\circ\text{C}$  are shown in Figure 3. The X-ray diffraction pattern demonstrates that with increasing pressure part of the low pressure phase transformed into an amorphous state (Fig. 2b). After 1.5 hours at 13.5 GPa and 850  $^\circ\text{C}$ , most of the starting material has transformed into the cubic-structured perovskite (Fig. 3b). As indicated by the images (Fig. 2a,3a), substantial length reduction occurred after phase transformation resulting largely from the high density of the new phase. With these directly measured sample lengths and travel times, the velocities during and after phase transformation can be calculated for the characterization of the phase transformation.

Elastic velocities as a function of pressure and temperature can be subsequently obtained by conducting measurements along multiple heating/cooling cycles when pressure is slowly released. The change in length of a  $\text{CaSiO}_3$  perovskite-phase sample as a

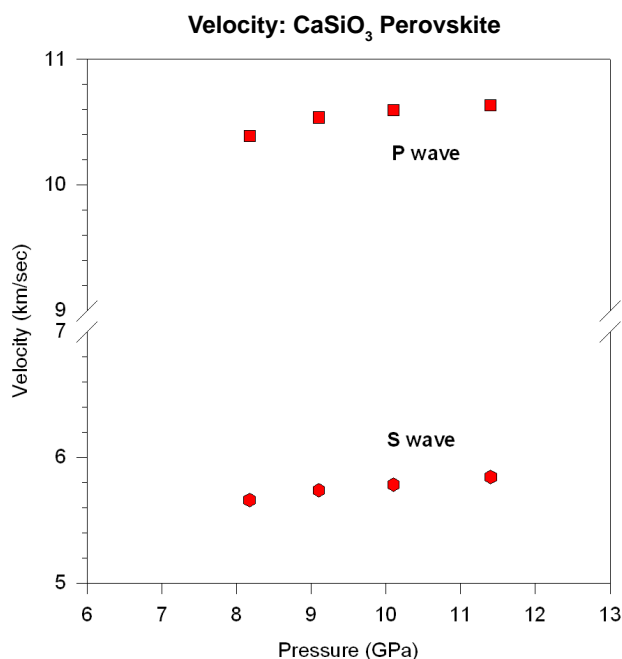
function of pressure at room temperature during decompression is shown in Figure 4. An abrupt increase in length occurred when the pressure was less than 1.8 GPa, indicating the breakdown of the meta-stable cubic phase. The X-ray diffraction taken at 1.8 GPa showed that the sample indeed started reverting back to its low pressure phase. Again, this feature can only be quantitatively captured with direct length measurement. Combining the sample length data in Figure 4 with travel times measured at the same pressure and temperature conditions, calculating P and S wave velocities is straightforward. Velocity data for  $\text{CaSiO}_3$



**Figure 3.** Image and X-ray diffraction pattern for  $\text{CaSiO}_3$  at 13.5 GPa and 850  $^\circ\text{C}$ . after phase transformation. Perovskite phase hkl's are shown in red.



**Figure 4.** Length change as a function of pressure obtained on decompression after the sample transforms into cubic  $\text{CaSiO}_3$  perovskite phase.



**Figure 5.** P and S wave velocities of  $\text{CaSiO}_3$  perovskite from 8 to 11.8 GPa.

perovskite in the pressure range of 8 to 12 GPa are summarized in Figure 5. The P wave velocity at 10 GPa compares well with theoretical data of Karki and Crain (1998), but the S wave velocities differ by 10%. The discrepancy cannot be reconciled until more experimental data becomes available. We plan to conduct experiments at higher pressures in order to eliminate uncertainties caused by minor untransformed low pressure phases that is beyond the detectability of the X-ray diffraction technique.

In summary, we have successfully adapted X-radiography for direct determination of sample length at high pressures and high temperatures in our ultrasonics and X-ray diffraction set-up. With combined measurements of travel time, X-ray diffraction, and sample length, elastic wave velocities of solid materials at high pressure and high temperature can be determined precisely. This technique opens new opportunities for quantitative studies of elastic property changes associated with phase transformations, rheological properties of materials at high pressure and high temperature, and elastic properties of multi-phase composites.

## References

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